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13. ABSTRACT (Maximum 200 words)

A unique experimental device, the Nanoworkbench, has been constructed and is being extensively tested at present. This device involves 4 independent STM tips which may be imaged by an SEM in order to locate their relative positions. These 4 STM probes will be used to make 4-point probe electrical conductivity measurements on nanometer objects. In addition, provision is made for Auger spectroscopy measurements on individual 10 nm objects which are being probed by the STMs. The instrument works in a number of initial tests. Preliminary examples of tests and measurements with this instrument are included in this report.

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Sincerely,

John T. Yates, Jr.

FINAL REPORT - FY 2000 ARO DURIP
PROJECT "Nanoworkbench for Analysis, Manipulation, and Excitation of Individual Nanostructures."

August 6th, 2001

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Summary

The nanoworkbench project has been funded by ARO/DURIP in two stages, partially in March 1999 and partially March 2000. The construction of this novel system includes a unique multiple tip STM device, coupled with a SEM and with an Auger analyzer for the study of single nanostructures, and a multiple UHV chamber system including molecular beam epitaxy (MBE) and multiple surface analysis capabilities.

Two students and Dr. Joachim Ahner are working full time on this project. The activities of the last two years have been mainly focused on the construction and the testing of the novel device. The construction is now almost completed and first results have been obtained. The report focuses on the following activities:

- Designing and building of a vibration isolated multiple chamber ultrahigh vacuum system, including a special sample transfer system.
- Development of vibration-isolated, variable temperature four tip STM.
- Exploration of a novel nano-MBE system.
- First results on directed growth of Ge/Si quantum dots and single wall carbon nanotubes.

1. Technological progress

1.1 UHV chamber system

The design of the three UHV chamber system shown as shown Figure 1 is completed and most of the vacuum parts have been constructed by the local machine shop. All chambers are connected with each other through a special sample transfer system. This system allows ultra clean sample preparation and characterization. The multiple source MBE (molecular beam epitaxy) chamber has been equipped with several electron beam evaporators for precise deposition and doping of metals and semiconductors.

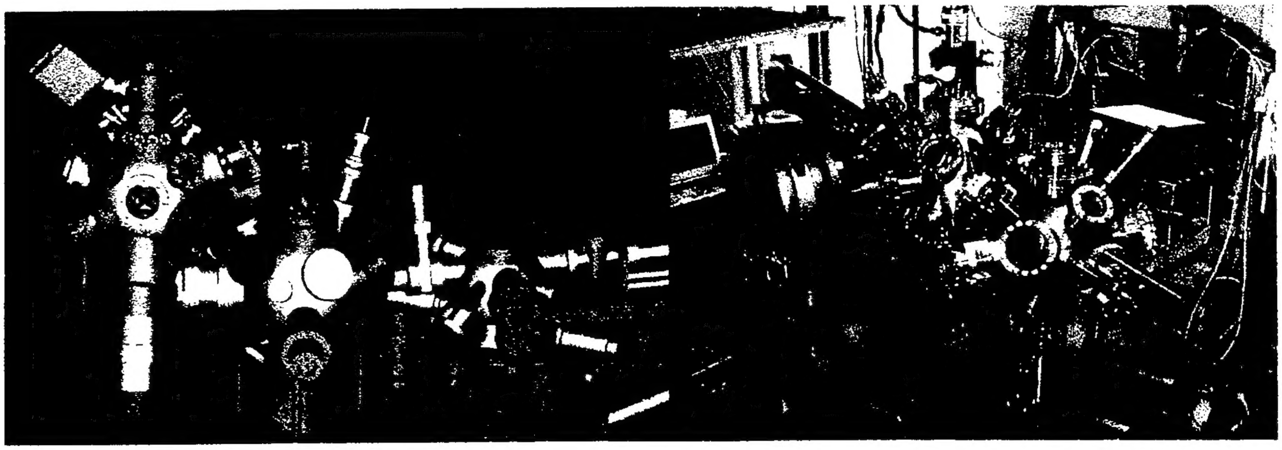


Figure 1. Three UHV chamber system. Left image: Drawing. Right image: Actual photograph.

1.2 Sample heating and cooling device

For the controlled growth of atomically flat and pure silicon surfaces, as well as for Ge and carbon directed self-assembled quantum dot structures it is important to have precise control of the sample temperature in each UHV chamber. For this purpose a special transferable sample box with integrated heating device has been build and tested (Figure 2). The temperature distribution was measured by an infra-red pyrometer and shows substantial derivations from the normally used thermocouple measurements. Sa,ple Heating in the upper central region is uniform to +/- 3 K.

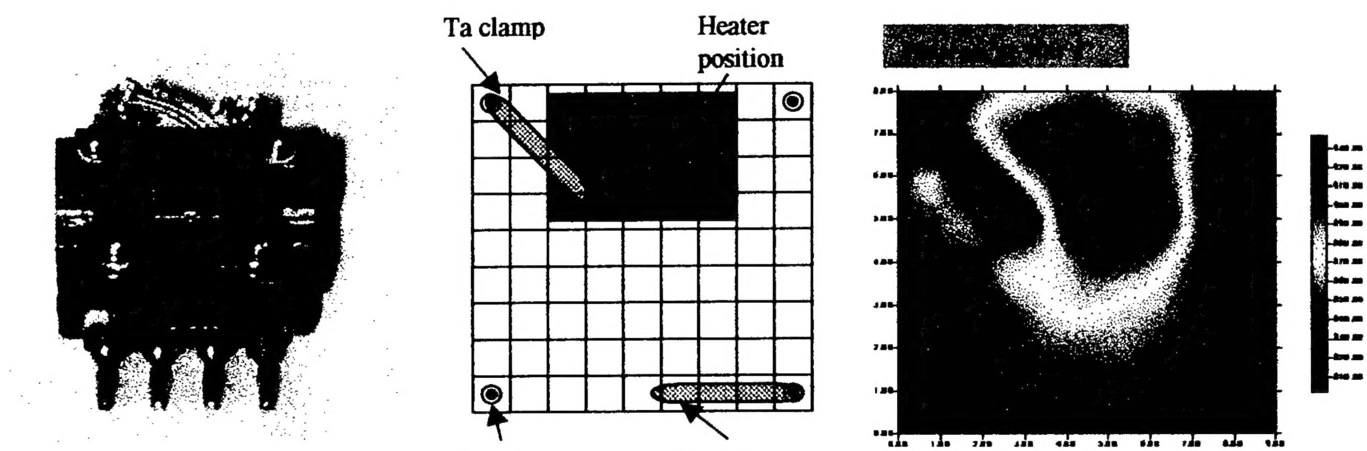


Figure 2: Left: transferable sample box with integrated heater and mounted Si (100) sample. Middle: Principle of pyrometer measurements matrix. Right: Temperature profile of the molybdenum plate, as measured with the pyrometer, for the home-made heater. The plateau value of 900°C is controlled via the thermocouple connections and LabVIEW automated computer control.

Because of the non-uniformity of the sample heating by this methode a second transferable heating stage has been designed in which ohmic heating of a thin Si(100) slice will be carried out.

1.3 **Multiple tip STM**

1.3.1 Novel nanomanipulator MM3

In collaboration with Kleindiek Nanotechnik, we developed the novel STM-nanomanipulator MM3, which has a large operation range and allows easy STM tip exchange in UHV. The MM3, as shown in Figure 3, combines one linear motion with two rotational motions of more than 180^0 of freedom.

State of the art electronics and appropriate software have been developed to allow precise macroscopic movement of the tip as well as STM imaging with atomic resolution. We consider this achievement as a milestone in the development of a nanoMBE system for Ge quantum dots growth, as described in section 2.

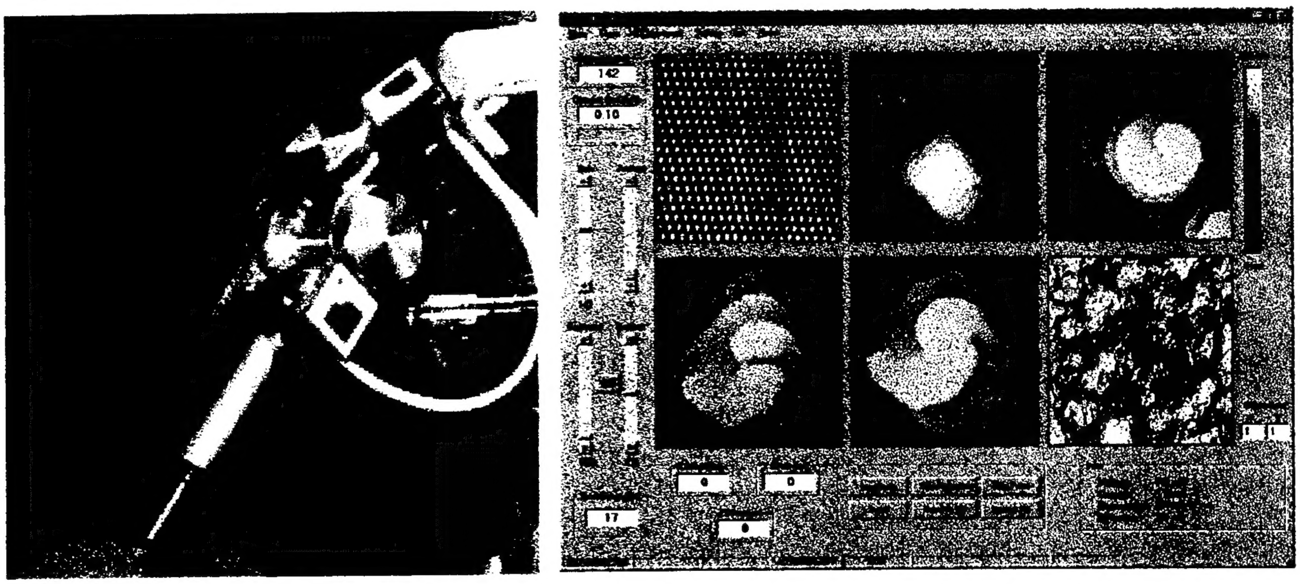


Figure 3. *MM3 nanomanipulator. A specially developed computer program can rotate the STM tip in any direction and atomic resolution can be achieved by imaging a plane tilted in any direction. The panel images on the right hand side show STM images of a highly ordered graphite sample taken with a nanomanipulator at air conditions. The upper left image demonstrates atomic resolution at a scan range of 8 nm x 8 nm. The other images show monoatomic steps at scan ranges between 100 nm x 100 nm to 50 x 50 nm.*

1.3.2 Four tip STM assembly

Figure 4 shows the homebuilt STM mounting assembly with 4 STM probes in place. The STM tips are mounted at 45 degrees to the axis, and at 90 degrees to each other. The actual photograph of the device (figure 4 center) shows additionally the recently developed high precision x,y,z-table with +/-10 mm range and sub-atomic resolution in all three directions. The table is equipped with novel position encoders (resolution better than 100 nm for the wide range course motion. The resolution in the fine motion is better than 0.01 nm). It is fully programmable by an electronic pattern generator and can be used itself as a scanner for four point STM measurements.

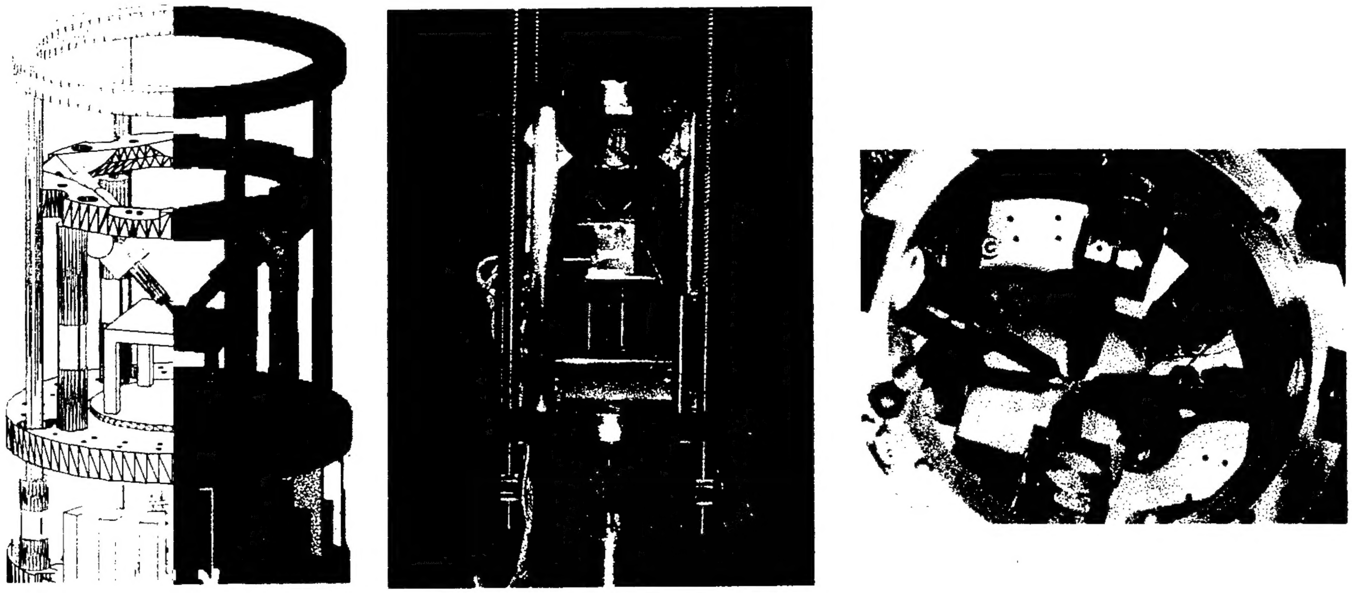
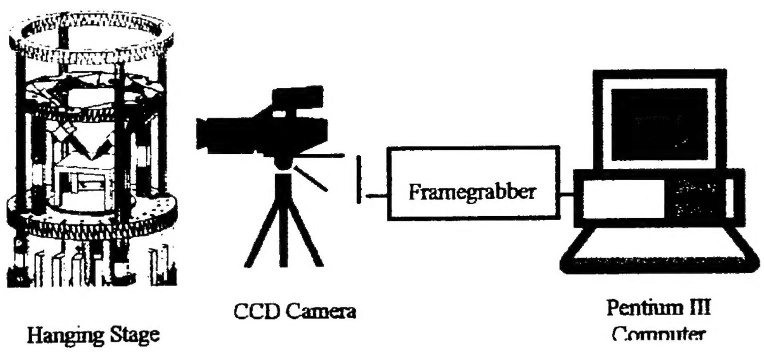


Figure 4. UHV multiple tip STM with eddy current damping system.Lleft hand: Design drawing,. Middle and right hand side: Actual device side and top view.

In order to isolate the STM system from external vibrations, springs have been fabricated by using Inconel 700 wire. All four springs have exactly adjusted force constants and together with an Eddy current damping system, coupling of the resonant frequencies of the apparatus is avoided.

1.3.3 Vibration analysis of Eddy current damping system of multiple tip STM



For high resolution multiple tip STM measurements an effective vibration isolation system is crucial. Figure 5 describes a vibration analysis experiment recently performed on the Eddy current damping system of the multiple tip STM. The results are shown in Figures 6 and 7.

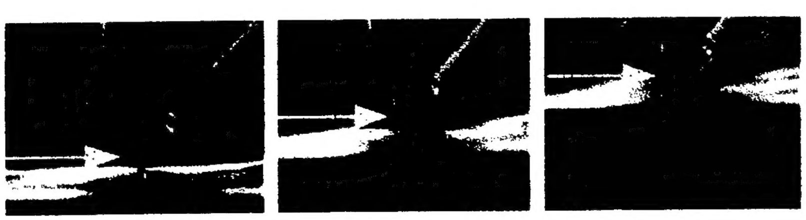


Figure 5. Setup for the vibration analysis experiment.

This analysis enabled us to optimize the force constant of the spring system as well as the damping constant.

The transmission function of the optimized system was measured (figure 7) and the resonance frequencies of the vibration isolation system (~1.5 Hz) could be kept far from the lowest natural frequency of the nanomanipulators (~200 Hz), leading to a very effective vibration isolation.

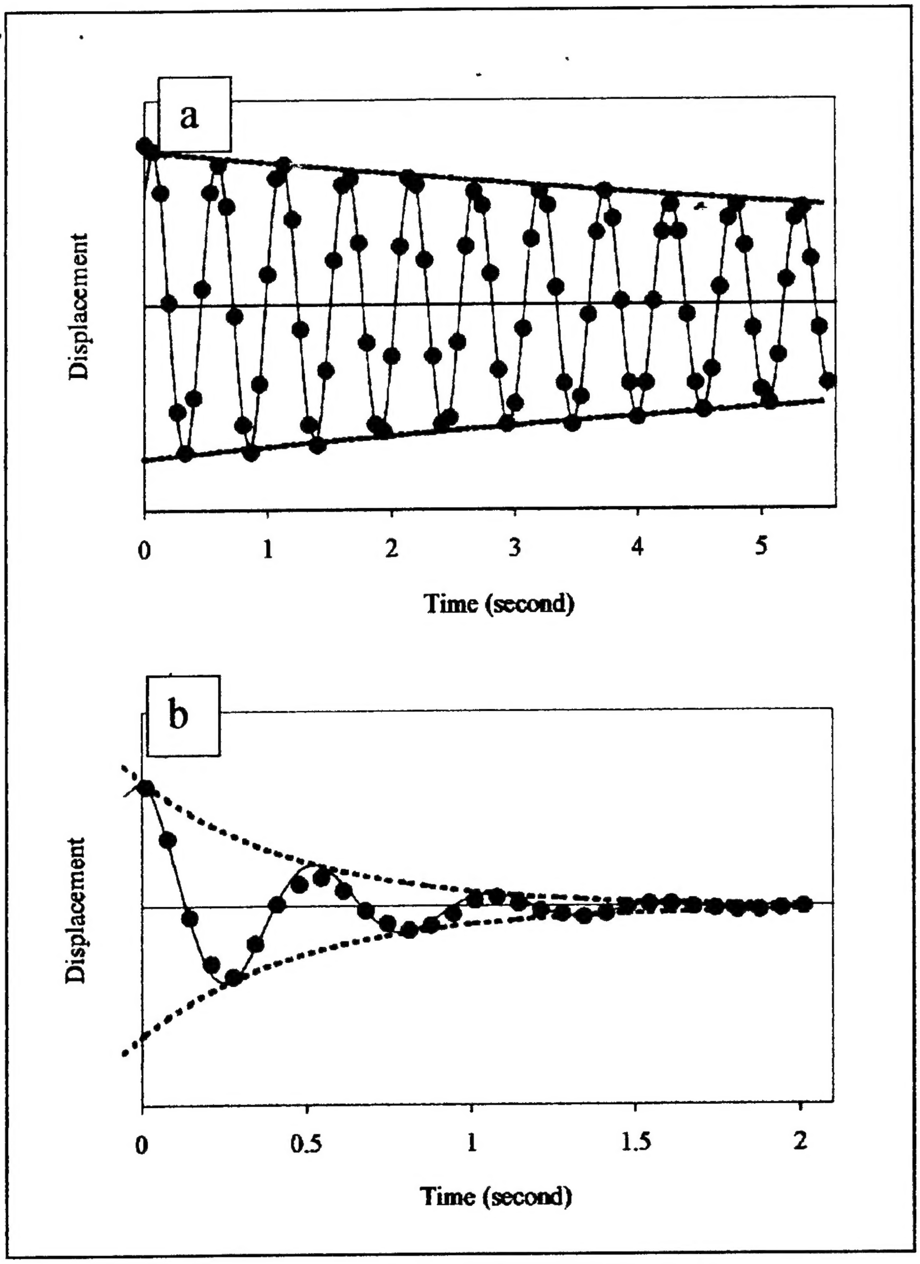


Figure 6. *Relaxation of the STM unit, (a) without the eddy-current damper and (b) with the Eddy-current damper.*

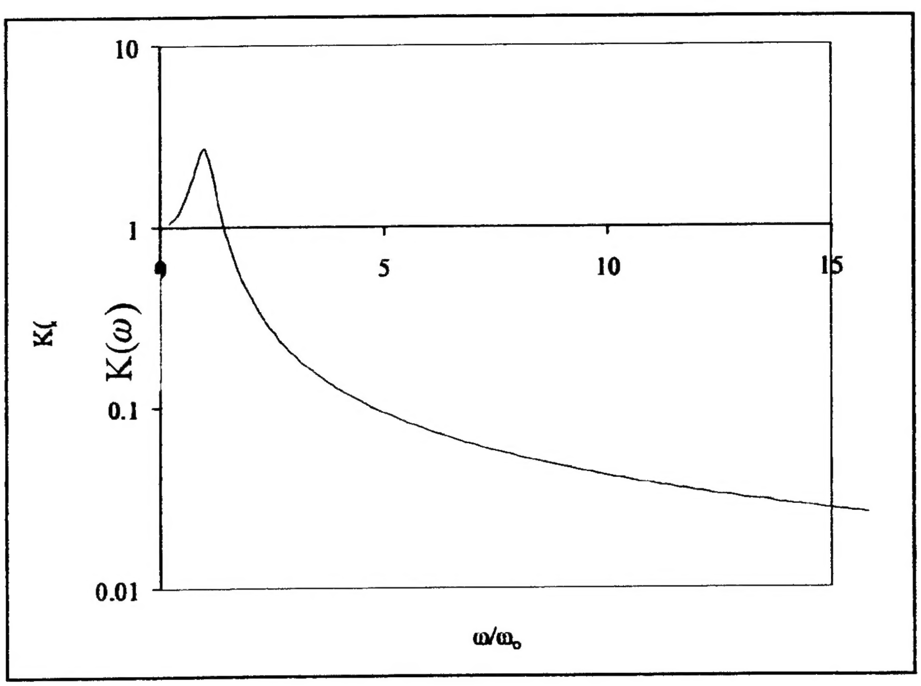


Figure 7. *Transfer function of the suspended multiple tip STM device. Inadvertent vibration amplification of a factor of 2.68 is expected at the natural frequency.*

1.3.4 Active vibration damping system

While the vibration problems effecting the multiple tip STM measurements have been solved by the internal Eddy current damping system, a separate solution had to be found to isolate the combined system SEM/STM) from external vibrations. This has been achieved with a six point active vibration isolation system for the whole UHV system. A special aluminum frame exhibiting a resonance frequency higher than 200 Hz has been designed and constructed in our local machine shop (figure 8). The active vibration isolation elements are located at the four corners and at the midpoint of the longer horizontal element.

***Figure 8**: Special frame exhibiting high resonance frequency. Together with a six point active vibration damping element it allows effective vibration isolation of the whole UHV system.*

2. First results

2.1 UHV- SEM. Manipulation of nanometer scaled material under SEM control

The operation of the FEI column was tested in UHV by using a home-build channeltron detector which was operated in the pulse-counting mode. Figure 9 shows the electron column mounted in the UHV chamber together with two STM tips. In the lower panel the two tips are shown with their reflected images in the smooth test surface.

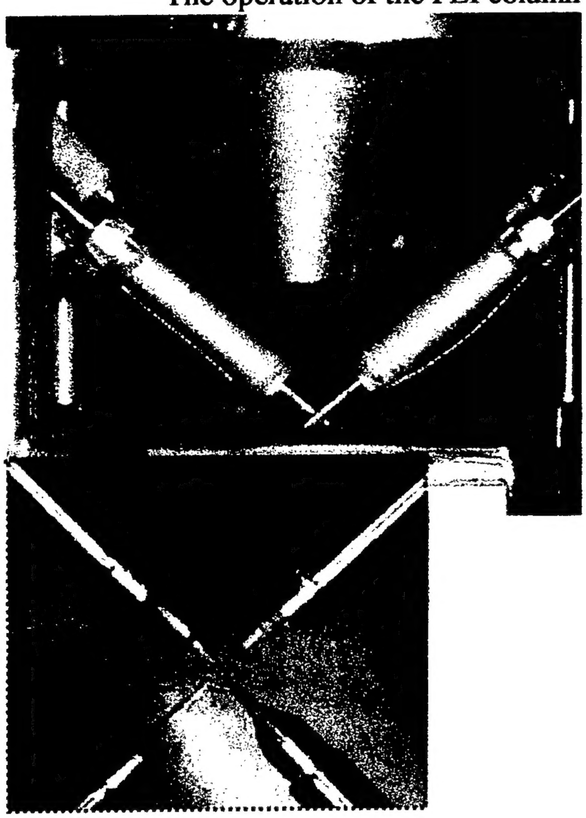


***Figure 9**. The electron column mounted in the UHV chamber (only the exit end is seen) together with the two STM tips, approaching each other.*

Figure 10 shows two SEM images from a video documenting a test where two STM tips approach an isolated gold coated polystyrene (PST) particle of 500 nm diameter. We succeeded for the first time to precisely position two tips under SEM control on selected PST particles in order to measure their electrical properties. This is an important step forward to explore novel nano-eletronic devices, as proposed.

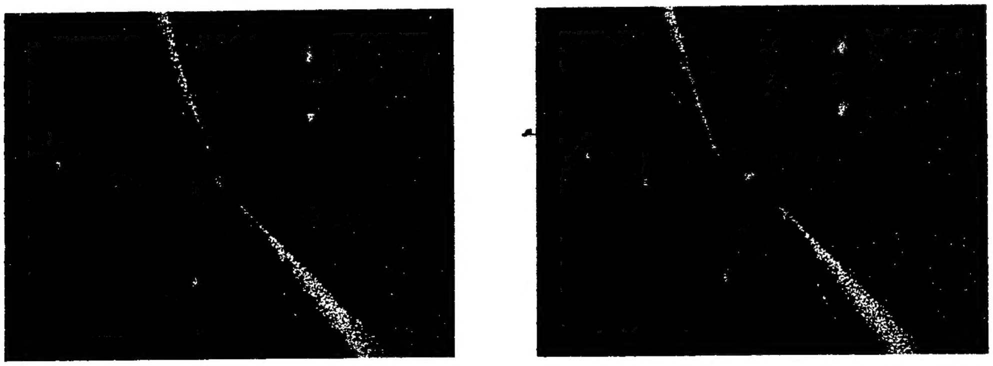


Figure 10. In-situ SEM imaging of two STM tips approaching an isolated PST particle. This images demonstrate for the first time the operation of a multiple tip STM..

2.2 Development of nano-MBE system for Ge quantum dot growth

Recently published results [1,2] indicate that Ge quantum dots grow on Si with a much smaller size after pre-deposition of carbon or Sb atoms. This ultra-small Ge dots show much enhanced photo luminescence than those grown without pre-deposition. Figure 11 shows on the right hand side carbon induced ultra-small Ge quantum dots. Ge dots grown without carbon form pyramids about ten times larger in size than required (left) [1].

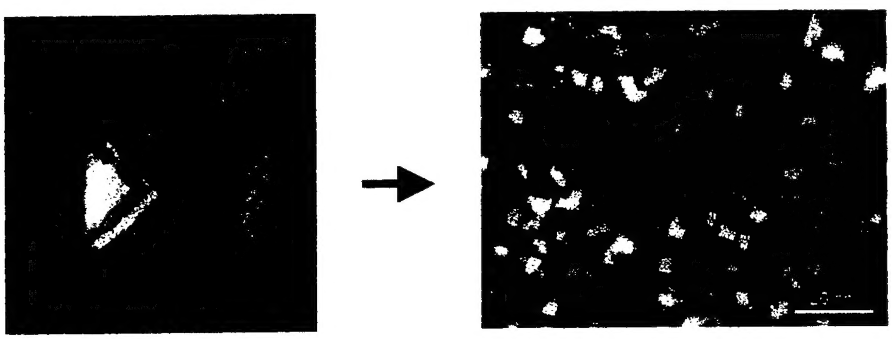


Figure 11: *Left: Typical Ge-dot grown on Si substrate with a diameter of 80 nm (too large for quantum electronic devices). Right: Precise control over carbon deposition results in the growth of ultra-small Ge quantum dots.*

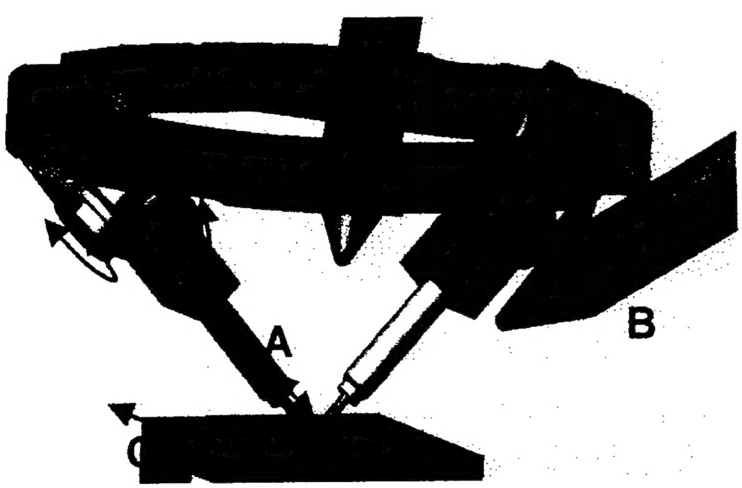


Figure 12. *Up to four independently working STMs (A) are combined in the nanoworkbench. They will be used to create patterns of carbon or Sb atoms on the Si substrate Evaporators(B) allow parallel deposition of Germanium and Si material. A high precision sample translation stage allows large scale positioning in x/y directions of the silicon substrate (C).*

Based on this information we are currently developing a NanoMBE system which uses the multiple tip STM (figure 12) to control the deposition of various atoms at controlled surface sites This atoms subsequently act as nucleation centers for the Ge quantum dots. We are working on a method to equip the tips with reservoirs of the specified atoms. From these reservoirs the atoms will diffuse to the tip end. For the deposition in UHV we will use field evaporation of atoms from the tip by applying relatively large voltage pulses or by a controlled tip approach as demonstrated in Figure 13.

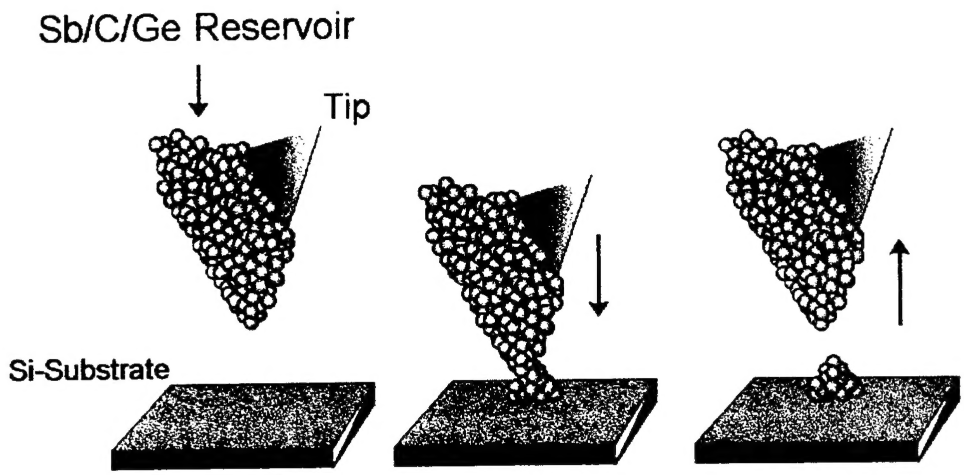


Figure 13. *Schematic diagram of the suggested mechanism of material transfer from the STM to the Si substrate, caused by an appropriate tip approach.*

2.2 STM tip assisted growth of Ge dot using a precursor molecule. Article published [3]

We succeeded in growing small Ge dots by using a precursor molecule and the STM tip. In Figure 14 the controlled growth of a Ge quantum dot in field-induced mode from Ge_2H_6 precursor molecules on the Si(111)-(7x7) surface is demonstrated. In this work the sample is biased negatively, and the emitted electrons cannot gain sufficient energy to induce electron attachment effects in the region occupied by the adsorbed Ge_2H_6 molecules. Thus the growth of the quantum dot is stimulated by the high electric field at the tip [4].

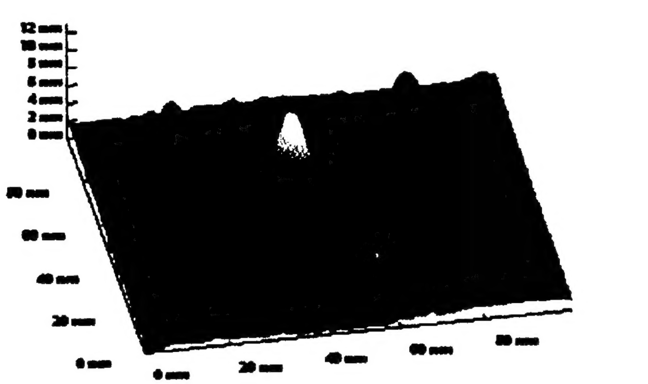


Figure 14. *3D constant current STM image ($V = -2.5 V$, $I = 0.01$ nA) of a typical GeH_X nanostructure grown at negative sample bias voltage. Growth parameters were: $V = -7$ V, $I = 0.01$ nA, $F = 4.1\times10^{12}$ molecules/cm^2s, $t = 3$ min.*

The experiments were conducted in a UHV chamber (base pressure ~ 6×10^{-11} Torr) equipped with a STM (Omicron) using electrochemically etched tungsten tips (R_{Tip}~ 5 nm). The nanostructure was grown at room temperature by dosing Ge_2H_6 gas through a conductance-calibrated stainless steel doser located 4 cm away from the tunneling junction, as schematically shown in figure 15. The flux of Ge_2H_6 is 4.1×10^{12} molecules/cm^{2}s.

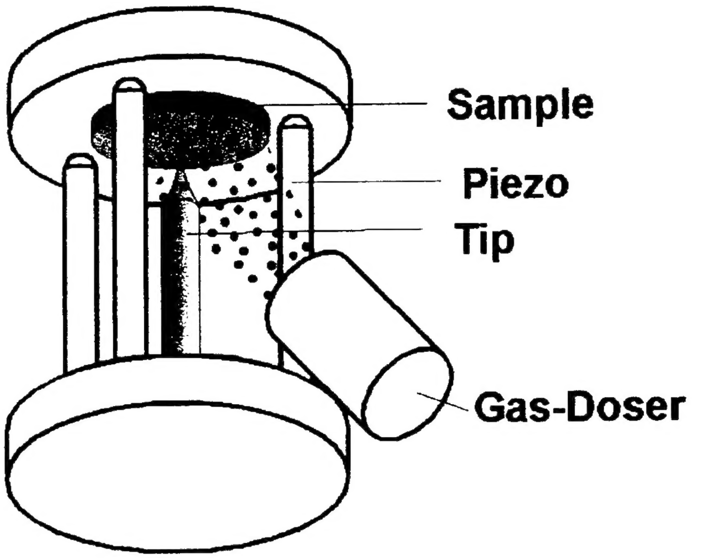


***Figure 15:** Schematic of experimental set-up for STM assisted growth of Ge dot.*

The STM was operating in a tunneling or a field emission regime (negative sample bias) in the constant current mode. Before the growth, the Si(111)-(7×7) surface has been saturated with Ge_2H_6.

The grown nanostructure in this preliminary experiment is likely to be of composition GeH_X rather than pure Ge based on studies of CH_3SiH_3 decomposition on Si(100) induced by electrons [5]. For useful Ge quantum dots, it is likely that subsequent annealing of these structures will be necessary.

2. Progress in growing isolated single wall carbon nanotubes for electronic devices.

In order to measure accurately the electronic transport properties of carbon nanotubes (CNT) of various diameters by using the multiple tip STM it is necessary to control the growth CNTs and suspend them on insulating pillars. One successful method is to use CVD on Ni or FeO_2 nanoparticles as catalysts [1-4] for the growth of aligned carbon nanotubes. For this purpose a CVD set-up according to the literature [2] has been built in our laboratory. We succeeded in growing free standing carbon nanotubes on various substrates as shown in figure 16a for a Ni-covered copper substrate. We also succeeded in growing single-sized Ni clusters with a diameter of approximately 10 nm on a Si (100) surface which subsequently act as catalysts for the carbon nanotube growth as shown in figure 16b.

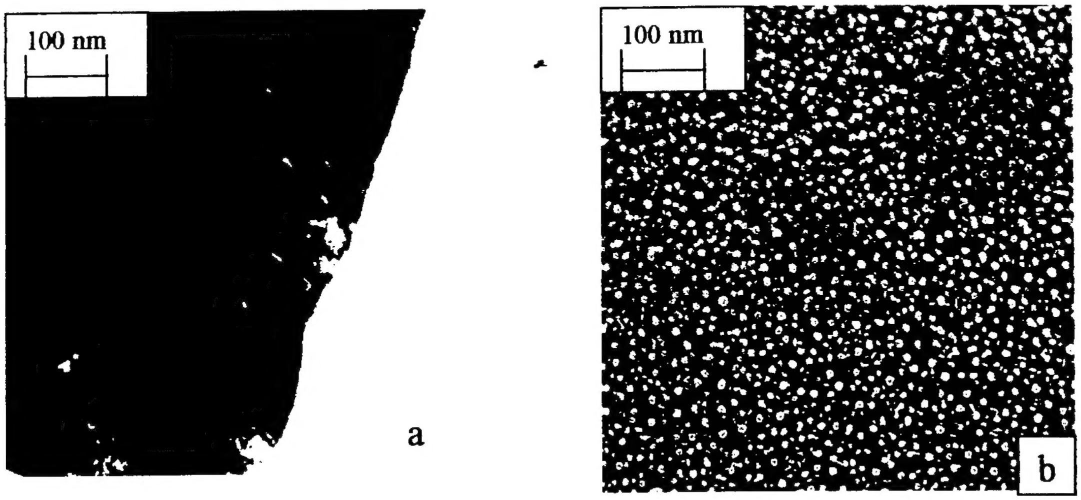


Figure 16: (a) Free-standing single carbon nanotubes grown on Ni catalysts deposited on a Cu substrate. (b) Uniformly-sized Ni particles grown on a Si(110) substrate.

4. Publications related to the project

- Joachim Ahner and John T. Yates Jr., "Novel Multiple-Tip STM and Nanoworkbench for Manipulating, Imaging and Analyzing of Nanostructures", Proceedings of the VW-Symposium "New Trends in Physics, Chemistry and Biology with Single Molecules", Wiesbaden, Germany, July 1999.

- J. Ahner, "Nanoworkbench for imaging, analysis, manipulation and excitation of individual nanostructures", MSTnews, 4, 16 (1999).

- Mawhinney, D.B., et al., *Infrared spectral evidence for the etching of carbon nanotubes: Ozone oxidation at 298 K.* Journal of the American Chemical Society, 2000. **122**(10): p. 2383-2384.

- Kuznetsova, A., et al., *Enhancement of adsorption inside of single-walled nanotubes: opening the entry ports.* Chemical Physics Letters, 2000. **321**(3-4): p. 292-296.

- Kuznetsova, A., et al., *Physical adsorption of xenon in open single walled carbon nanotubes: Observation of a quasi-one-dimensional confined Xe phase.* Journal of Chemical Physics, 2000. **112**(21): p. 9590-9598.

- S. Mezhenny, I. Lyubinetsky, J. Levy, and J. T. Yates, Jr., "STM tip assisted growth of Ge dot using a precursor molecule", *J. Vac. Sci. Technol.* B **19**, 567 (2001).

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